Reduction of Oxygen Transport Resistance in Polymer Electrolyte Fuel Cell by Modifying Ionomers

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論 文 名　：Reduction of Oxygen Transport Resistance in Polymer Electrolyte
　　　　　　Fuel Cell by Modifying Ionomers
　　　　　　（アイオノマー種に着目した固体高分子形燃料電池内の酸素拡散抵抗の
　　　　　　低減）

区　　　分　：甲

論 文 内 容 の 要 旨

　　　In this work, hydrogen energy is focused on as one of the solutions for global environmental problems through the combination with renewable energy. Polymer electrolyte fuel cell (PEFC) generating electricity with a hydrogen fuel is particularly studied among technologies related to hydrogen energy. Although fuel cell vehicles (FCVs) are already commercialized. PEFC has recently been paid more attention to for heavy duty vehicles (HDVs) like buses or trucks. Regardless of the technology developed for FCVs, the operation at higher current density is further necessary for HDVs. Current PEFC operation is limited due to high oxygen transport resistance (OTR) in the cathode catalyst layer. In the cathode catalyst layer, the ionomer covering Pt catalyst surface, which is essential for proton conduction, leads to oxygen transport loss. This loss especially becomes a serious problem at higher current density where the reaction is transport limited. With low Pt catalyst loading, OTR through the ionomer is more pronounced because more oxygen must be delivered to the smaller Pt surface. Therefore, reducing OTR in the cathode catalyst layer is important for next generation PEFC, which is directed toward low cost HDV application.

　　　In this study, in order to tackle the issue with OTR in the cathode catalyst layer, reduction of OTR was aimed by a simple yet effective method. Since Nafion® is commonly used as a standard ionomer, high oxygen permeable polymer was blended with Nafion® in certain mass fractions, and then applied to the cathode catalyst layer to reduce OTR. OTR around Pt catalyst (R_{Pt}) was separately analyzed from OTR by passing through the pores in the cathode catalyst layer (R_{pore}), and effectiveness of blend ionomers on OTR was fully discussed.

　　　In Chapter 1, research background and future direction with critical issues of PEFC were introduced. After narrowing the focus on OTR, previously reported research approaches toward reducing OTR were explained. Lastly, objectives and research approaches of this study were introduced.

　　　In Chapter 2, methods of preparation and characterization of blend ionomers were first explained. Then, methods of preparation and evaluation of PEFC with blend ionomers were discussed. Particularly, analytical procedures of OTR was fully explained.

　　　In Chapter 3, Poly{1-phenyl-2-[p-(trimethylsilyl)phenyl]acetylene} (PTMSDPA) was

chosen as a high oxygen permeable polymer. Synthesized PTMSDPA was fully characterized. By blending PTMSDPA into Nafion® with different fraction (1.25%, 2.5%, 3.75%, 5.0%), the blend ionomers were applied to the cathode catalyst layer. Although activation overvoltage was rather increased owing to decreased proton conductivity derived by reduced hydrophilic channel within the ionomer. However, improved performance at high current density was observed. When 2.5% PTMSDPA was blended into Nafion®, the performance at high current density resulted in maximum. When R_{Pt} was separately analyzed from R_{pore}, at low Pt loading like 0.1 mg/cm^2, R_{pt} was found to be significantly reduced. The 43% reduction was successfully done in comparison to the use of Nafion® only. Additionally, R_{pore} was also reduced owing to resulting larger macropores in the cathode catalyst layer. However, increasing the fraction of PTMSDPA was rather found to increase R_{pore} owing to hydrophilic properties of PTMSDPA, and importance of selection of more hydrophobic polymer was realized.

In Chapter 4, polymer intrinsic of microporosity (PIM-1) was chosen for blending because of relatively more hydrophobic character than PTMSDPA in addition to high oxygen permeability. By blending PIM-1 into Nafion® with different fraction (2.5%, 5.0%, 7.5%, 10%), activation overvoltage was reduced comparing to PTMSDPA blend ionomers owing to improved proton conductivity derived by well incorporation of PIM-1 into the hydrophobic domain of Nafion® ionomer. When 5% PIM-1 was blended into Nafion®, best performance at high current density was observed through the reduction in both R_{Pt} and R_{pore}. When Pt loading was reduced like 0.1 mg/cm^2, 50% reduction of R_{Pt} was successfully done in comparison to the use of Nafion® only. Furthermore, R_{pore} was reduced by 11% owing to the resulting further increased pore size in the cathode catalyst layer and also less hydrophilic character of pore surface.

In Chapter 5, general conclusion along with new findings of this research was summarized, and future outlook for further reducing OTR was discussed.